報告內容

1. 前言

PCDD/Fs, known as dioxins, are among the most toxic chemical compounds in the environment. Generation and release of PCDD/F's from anthropogenic activities have raised tremendous concerns due to acute and chronic health effects such as immune, nervous, endocrine, reproductive, and carcinogenic potential of dioxin exposure.

Waste incineration and burning is known as one major source of dioxin emission (Ma, Lai, & Chan, 2002; Pulles et al., 2006). In Taiwan, incineration is becoming a dominant municipal waste treatment method. It is estimated that more than 90% of municipal wastes (over 9 million metric tons per year in quantity was treated by incineration in 2003 (Ma, Lai, & Chan, 2002). It is therefore important to investigate the dioxin exposure due to incinerators in Taiwan.

2. 研究目的

The purpose of the project is to develop statistical methodology in spatiotemporal distributions of environmental monitoring data. The data set analyzed in this project is airborne dioxin concentration measurements near a local incinerator in central Taiwan. The main objective is to find whether there was relationship between dioxin concentration measurements and the incinerator. Also, one other goal of the study is to investigate the spatial distribution of lifelong risk of residents living in the vicinity of the incinerator.

3. 文獻探討

Although atmospheric particle size distributions near an incinerator has been analyzed (Chao, et al), and transfer of dioxin risk between waste incinerators in Taiwan has been studied, the relationship between dioxin concentration and distance to corresponding incinerator has not been fully investigated statistically. This study provides a formal statistical analysis of spatiotemporal dioxin concentration distributions in the vicinity of a local incinerator in Taiwan.

4. 研究方法

A straight line across the center of the incinerator with the angle ω between the horizontal line representing the wind direction was applied to each of the seasonal collected samples. Then $\cos\theta$ represents the relative contribution of the wind direction to the PCDD/F concentration of the sampled site, where θ $\left(0 \le \theta \le \pi\right)$ is the angle between the lines connecting the sampling site and the incinerator and the wind direction. For the upwind sampling sites, $\cos\theta$ is negative ($\pi/2 \le \theta \le \pi$), and the PCDD/F's concentration is expected to be less than normal owing to the wind. The effect on PCDD/F's concentration is otherwise for the downwind sampling sites. A linear regression model was applied to the logarithms of dioxin concentrations of the sampling sites with explanatory variables *distance* and $\cos\theta$, where *distance* denotes the distance between the site and the incinerator. Since the wind directions varied during the sampling seasons, and meteorology records were available only at sites F and G, the angle ω was rotated every 10 degrees to find the best fitted regression model with the optimal R^2.

Since there were only seven observations for each season's sample, results based on linear regression model may not be reliable. In addition, the observations at different sites are spatially correlated with distributions unknown. Thus, the model assumptions of normality and independent identical distribution may not hold. To assess the association of the dioxin concentrations with the distances to the incinerator and wind directions in void of model assumptions, one may employ the nonparametric correlation index Kendall's tau τ. Let (Z_i, H_i, W_i) be the triple measurements associated with sampling site $i, i = 1,\dots,n$, where Z_i is the dioxin concentration measurement, H_i is the distance to the incinerator, and W_i is the wind direction contribution $\cos\theta$. For the association between concentration and distance, let C (D) be the number of pairs of observations such that the order of their concentrations is concordant (discordant) with the order of their distances. That is,

$$C = \sum_{i \ne j} I\{(z_i > z_j) \wedge (h_i > h_j)\},$$

$$D = \sum_{i \ne j} I\{(z_i > z_j) \wedge (h_i < h_j)\},$$

where $I\{\cdot\}$ is the indicator function with value 1 if the logical statement in the parenthesis is true, and 0 otherwise. The Kedall's tau is defined as $\tau(Z,H) = (C-D)/(C+D)$. The correlation coefficient τ is by definition between -1 and 1. The closer to the maximum (minimum) value 1 (-1) the more evident that there is positive (negative) association between Z and H. If the number of observations n is large enough, then asymptotically τ is normally distributed, and statistical significance of the association can be described by the corresponding p-value from the normal distribution. However, there were only seven observations from the study sites, asymptotical results do not apply in this case. One way to find the statistical association in the case of small sample size is the nonparametric permutation test. Under the assumption that the observations are interchangeable, the observations of Z and H can be relocated to have a total of $7! = 5040$

permutations. The exact p-value of the observed τ can then be calculated by matching its order among the 5040 τ 's from smallest to largest after the permutations. The permutation test for the association between Z and W can be calculated similarly.

The associations between the concentration Z, the distance to the incinerator H, and the wind contribution W, are in fact inter-correlated. For example, the dioxin concentration at downwind site is expected to be higher than that at the upwind site, even though its distance to the incinerator may be greater than the latter. Therefore, one may need to adjust the association index Kendall's tau presented above by controlling on a third variable. For the association between Z and H conditioning on W, consider a pair of observations (z_1, h_1, w_1) and (z_2, h_2, w_2). If conditioning on $w_1 > w_2$, we have $(z_1 < z_2) \wedge (h_1 > h_2)$, then there is more evident that the difference in concentration is due to distance. Let the adjusted concordant (discordant) pairs conditional on W to be

$$C^* = \sum_{i \neq j} I\{(z_i < z_j) \wedge (h_i < h_j)\} I\{w_i > w_j\},$$

$$D^* = \sum_{i \neq j} I\{(z_i < z_j) \wedge (h_i > h_j)\} I\{w_i > w_j\}.$$

The Kendall's tau conditional on W is $\tau^*(Z, H \mid W) = (C^* - D^*)/(C^* + D^*)$. Similarly, the Kendall's tau $\tau^*(Z, W \mid H)$ conditional on H is defined with

$$C^* = \sum_{i \neq j} I\{(z_i < z_j) \wedge (w_i < w_j)\} I\{h_i < h_j\},$$

$$D^* = \sum_{i \neq j} I\{(z_i < z_j) \wedge (w_i > w_j)\} I\{h_i < h_j\}.$$

The permutation test for the adjusted Kendall's tau can be performed similarly as before, conditioning on the discordant pairs of (Z, W) for $\tau^*(Z, H \mid W)$ and concordant pairs of (Z, H) for $\tau^*(Z, W \mid H)$.

A hierarchical spatial model with response variable the logarithm of the dioxin concentration $Y(\mathbf{s}) = \log Z(\mathbf{s})$ is assumed as follows:

$$\mathbf{Y}(\mathbf{s}) \mid \mathbf{X}(\mathbf{s}) \sim N(\mathbf{X}(\mathbf{s})\boldsymbol{\beta}, \sigma^2 \mathbf{H}(\phi)),$$

where $\mathbf{Y}(\mathbf{s})$ is the vector of the responses, $\mathbf{X}(\mathbf{s})$ is the matrix of the covariables, $\mathbf{s} = (\mathbf{s}_1, \ldots, \mathbf{s}_7)'$ is the vector of the sites' coordinates, and $\sigma^2 \mathbf{H}(\phi)$ is the covariance matrix with $H(\phi) = \left(\rho\left(\left\|\mathbf{s}_i - \mathbf{s}_j\right\|, \phi\right)\right), \rho\left(\left\|\mathbf{s}_i - \mathbf{s}_j\right\|, \phi\right) = \exp\left(-\phi\left\|\mathbf{s}_i - \mathbf{s}_j\right\|\right)$. The implicit assumption of the model is that the spatial correlation is second-order stationary, that is, the correlation between two sites depends only on their distance with each other. The Markov Chain Monte Carlo (MCMC) simulations using the WinBUGS software may be applied for the estimation of the model parameters. For prediction of the response Y_0 at a new site $\mathbf{s}_0$ with associated covariate vector $\mathbf{x}(\mathbf{s}_0)$, the Bayesian kriging may be applied by sampling y_0 from its posterior distribution $f\left(y_0 \mid \mathbf{Y}(\mathbf{s}), \boldsymbol{\theta}, \mathbf{x}(\mathbf{s}_0)\right)$, where $\boldsymbol{\theta} = \left(\boldsymbol{\beta}', \sigma^2, \phi\right)'$ (Banerjee, Carlin, Gelfand, 2004, p133).

5. 結果與討論

It was found that the dioxin concentrations decreased with the distance to the incinerator, and were dependent on the seasonal winds. This result was obtained by nonparametric permutation test which requires no distributional assumptions at all. Therefore, it is quite plausible that municipal waste incinerators are responsible to environmental exposure of dioxins to a certain degree. However, since the permutation tests were only based on rearrangement of measurements from 7 points, further investigations needs to be done in the future.

參考文獻

張簡國平等（2001）“彰化縣委託操作管理溪州垃圾資源回收廠（焚化爐）環境監測報告書”九十年第一、二季報告書，彰化縣環境保護局

張簡國平等（2002）“高廠代彰化縣管理操作溪州垃圾資源回收廠（焚化爐）環境品質監測工作”九十一年第一、二季報告書，彰化縣環境保護局

張簡國平等（2002）“臺北市北投與木柵垃圾焚化爐戴奧辛與重金屬污染環境監測報告書”

陳德祐（2004）“臺中市垃圾焚化爐臨近區域空氣沉降戴奧辛濃度空間統計模式與分析”，淡江大學數學所碩士論文

翁仕旻（2005）中部地區某焚化爐附近地區空氣中戴奧辛暴露與風險評估，淡江大學數學研究所碩士論文。(Major outcomes of the proposed project)

Banerjee, S., Carlin, B. P., Gelfand, A. E., 2004. *Hierarchical Modeling and Analysis for Spatial Data*. Chapman & Hall/CRC, Boca Raton, Florida.

Chao, M.-R., Hu, C.-W., Ma, H.-W., Chang-Chien, G.-P., Lee, W.-J., Chang, L. W., Wu, K.-Y. (2003) Size distribution of particle-bound polychlorinated dibenzo-p-dioxin and dibenzofurans in the ambient air of a municipal incinerator. Atmospheric Environment 37, 4945-4954.

Ma, H.-W., Lai, Y.-L., Chan, C.-C. (2002) Transfer of dioxin risk between nine major municipal waste incinerators in Taiwan. Environment International 28, 103-110.
Pulles, T., Kok, H., Quass, U., 2006. Application of the emission inventory model TEAM: Uncertainties in dioxin emission estimates for central Europe. Atmospheric Environment 40, 2321-2332.

計畫成果自評

The research outcomes of the project conform the primary goal of the proposal. Since similar findings have not been discussed in the literature, and it was obtained by rigorous statistical analysis, the results are publishable in major environmental science journals. We are currently writing a manuscript to be submitted to *Atmospheric Environment*.